

SHOCK TUBE SPECTROSCOPY : REVIEW AND RECENT RESULTS

by

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ABSTRACT

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Conventional shock tubes are used in the temperature range 5,000-15,000°K for measuring atomic transition probabilities ("oscillator strengths"). Reduction of line intensities to atomic constants proceeds via knowledge of gas conditions. Results for Cr I and Cr II lines are discussed, based on experiments in the Michigan shock tube. Experience with this instrument is reviewed regarding measured pressures, temperatures and flow-durations. Properties of the new Maryland shock tube give greater flexibility for spectroscopic work. An improved flash-lamp allows temperature measurements at least up to 15,000°K by means of spectral line-reversal. This and other techniques will provide simultaneous and independent state measurements, so that shock tube theory can be checked rather than be relied upon in the determination of oscillator strengths.

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1. General Discussion

During the past ten years, various types of shock tubes have increasingly been used as light sources for atomic and molecular spectroscopy. Principal attention has been given to measurements of line-broadening parameters and radiative transition probabilities. Conventional (i.e., gas driven) tubes operate up to 15,000°K, explosively driven tubes somewhat higher, and electrical tubes up to 50,000°K easily. The interest in all of these stems from what one might call the "hyperadiabatic" heating of material induced by its passage through a shock front. The temperature ratio across a shock wave can be much higher than that for an isentropic process of the same pressure ratio, because of the pressure-density relations peculiar to shock-wave thermodynamics. This theory, in addition, predicts the gaseous state-variables from simple observables (such as the shock wave velocity) and says that the temperatures achieved may be systematically varied over a wide range by adjustments of the experimental parameters. So it was generally hoped

that atomic constants could readily be measured by correlating the observed spectra with the calculated temperatures, level populations, fractional ionizations, etc. Thus one could expect to get precise f-values for many atoms and ions and good line-shapes as functions of ion and electron density, and thereby have in hand those quantities necessary for understanding other light sources such as stellar atmospheres.

In a modified form, this view has been borne out, but the intervening years have shown that shock tube plasmas are not always

described by the simple Rankine-Hugoniot theory. This realization has come with the development of direct state-measurements on the hot gas, and now the prevailing view seems to be that such measurements are essential to any shock tube-spectroscopic program. In the general area of temperature measurements, for example, it has sometimes been observed with conventional tubes that flow durations are too short to allow complete relaxation to thermal equilibrium and that the "temperatures" for ionization and excitation may therefore be much lower than the gas-kinetic temperature.^{1,2} An opposite effect is sometimes encountered with electrical shock tubes, where the intense luminosity of the driving arc ionizes the gas ahead of the shock to an extent that the gas behind it, though in equilibrium, is much hotter than one would expect from the shock speed alone.^{3,4}

Since these and many other effects can occur (and since a complete theoretical accounting for them is not likely) the most fruitful procedure is to make several independent state-measurements of sufficient quality that the spectroscopic results have a clearly defined and useful precision, irrespective of comparisons with Rankine-Hugoniot theory.

These comparisons are still useful, however, from another point of view. We will give an example below of how such observations not only give clues as to plasma non-uniformity (a potential difficulty with all spectroscopic sources) but also indicate how those difficulties can be minimized. Thus we take the dualistic point of view that one should measure the hot gas conditions as completely as possible and continually set these results against compressible flow theory. In this way the experimenter gains both

the desired spectroscopic information and a better understanding of the processes in his spectroscopic source. The examples we will give of both procedures stem from our work^{5,6,7} with Turner's conventional shock tube⁸ at the University of Michigan in Prof. Otto Laporte's laboratory. We are currently using these and other methods at the University of Maryland. As before, we intend measuring oscillator strengths for lines of astrophysically interesting elements and finding line-broadening parameters wherever feasible.

2. Effects of Boundary Layers.

To illustrate the usefulness of comparisons with ideal shock tube theory, we consider first the customary wave diagram (Figure 1) for conventional tubes, with distance running to the right and time upwards. For strong shocks in the rare gases (with no ionization) the density ratio ρ_2/ρ_1 asymptotically approaches 4, and P_2/P_1 approaches 2.5 from below. Thus the hot, doubly compressed gas, whose spectrum is observed at far right, has a density roughly 10 times the value initially set in the right hand chamber. Hence its length should be no less than 1/10 the chamber length when the reflected shock and interface interact, no matter what the initial pressure P_0 .

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Now, one might be prepared for some deviations due to imperfect diaphragm breakage, but the actual performance is far worse, as indicated in Figure 2. This shows several wave-photographs of the reflection process with time now running to the right. With decreasing initial pressure p_0 , the length L_R (between the end wall and the interface interaction) decreases without limit. The cause of this is boundary layer growth on the tube walls and gas mixing, which impart an increasing three-dimensionality to the primary flow as the initial pressure is lowered. Though these effects

received some early attention at Toronto and Michigan, and by Toennies and Greene⁹ at Brown University, the first systematic study was made by Duff¹⁰ at Los Alamos using an electron-beam densitometer. Extensive studies have followed by Roshko¹¹ and Hooker.¹²

The obvious practical effect is to reduce the time of observation at any station and therefore to reduce one's ability to observe conditions which are fully relaxed to thermal equilibrium. A less obvious matter is what happens to the gross flow variables, particularly the gas density. One might naively ascribe this decreasing flow length to an increasing compression ratio, C_R , i.e., an anomalously high density across the entire flow cross-section. This is fortunately not the case, but it is true that high densities are developed in the wall boundary layer, and the hot gas can therefore be made highly non-uniform along any line of sight. Since this becomes worse with decreasing p_0 , one is clearly best off with the highest p_0 consistent with the desired temperature and attainable driver gas pressures.

We have also verified that these effects of decreased testing time and increased non-uniformity become worse with an increasing ratio of surface to volume. With small tubes operating at or below a mm (Hg) pressure, one must clearly be very careful about uniformity along the spectroscopic line of sight. For realistic aerodynamic experiments in the low pressure regime, large diameter tubes such as that used at Avco seem imperative. We have designed the new shock tube at Maryland around an initial pressure range of 1 to 10 cm (Hg). The cross-section is about 8 x 10 cm; being rectangular, it allows clear representations of wall boundary layers as with the Michigan tube (Figure 3). The luminosity in both Figures 2 and 3 arises not from ionization but rather a non-equilibrium

emission by the C_2 molecule arising from the pyrolysis of CH_4 (added to the neon carrier gas in amounts less than 1%). The operating range of the new shock tube yields thinner and almost wholly turbulent boundary layers and provides about 200 microseconds of steady conditions behind the reflected shock, near the tube's end wall.

3. Temperature Measurements

Having made such kinematic comparisons with hydrodynamic theory in order to find some operating conditions which seemed better than others, we had then to measure the state-variables directly to put a solid foundation under our spectroscopic results. The predicted pressures were confirmed¹³ to within 5% by means of quartz transducers having response times of order 3 microseconds. The most informative study for spectroscopic purposes was the measurement of electronic excitation temperature by means of spectral line-reversal.^{7,14,15}

Some line-reversal results are shown in Figure 4, where we have verified shock tube theory for the temperature behind the

reflected shock, over a range of primary shock strength (horizontal axis). These experiments involved 3/10% chromium carbonyl in neon in order to generate many lines of Cr I and Cr II and measure their transition probabilities. The theory predicts severe temperature depression relative to the case of pure neon, and this is borne out by these measurements. This temperature effect arises primarily from dissociation of the six CO groups coming out of each $Cr(CO)_6$ molecule. Using such volatile metal compounds, one gets the metal into the gas phase with known abundance; temperature measurements then specify the ionization and excitation equilibria, and the observed spectral line-intensities can then be converted into absolute atomic transition probabilities.

Some of the chromium lines are evident in the time resolved spectrogram of Figure 5, where we also see the varying continuum intensity due to the line-reversal lamp.¹⁶ Without such a lamp, these shock tube spectra show only faint continua under strong lines. Viewing the lamp through the shock tube gas, we see the gas in absorption when the flash lamp is "hotter" than the gas. At reversal, all spectral features disappear; the Planck function for the shock tube gas is put in evidence via absolute intensity calibration of the flash lamp. We find reversal at the same temperature for all spectral lines, regardless of their excitation potentials, which is to say that thermal equilibrium is achieved to within the 3% measurement error.

The results so far^{6,7} are that we have absolute transition probabilities (10% - 15% precision) for forty lines of Cr I and twenty lines of Cr II, which can now be turned to temperature and abundance determinations in other spectroscopic sources. In particular, our revision of the relative scale for Cr I appears to have cleared up a previous difficulty in interpreting Fraunhofer intensities in the solar spectrum.¹⁷

4. Conclusion

Our present and future experiments are built upon this preceding work we have briefly described. The new shock tube has just begun to operate and appears to fulfill its design, namely that the tube cross-section and useful initial pressures are sufficiently large that the flow fields are not grossly distorted by boundary layers and that steady conditions prevail long enough for thermal equilibrium to be obtained in most cases. The accessible temperature range is 5,000 - 15,000°K and therefore allows a range of spectroscopic studies from molecules and metallic atoms, at one end, to most single ions at the other. Similar work emphasizing metallic spectra continues at Michigan under Laporte and at Ohio State University under Slettebak. We plan to concentrate on the more abundant light elements such as carbon, oxygen and sulfur and to buttress the results with an increasing number of independent state-measurements. A new flash lamp is now in operation giving maximum brightness temperatures $\sim 20,000^\circ K$ and therefore allowing reversal temperature measurements over the entire anticipated range. An optical interferometer (Mach-Zehnder, 30 cm) is being modified for use with the new tube so that electron densities¹⁸ can be measured directly down to 10^{16} cm^{-3} (or total particle densities in cases of low ionization). Absolute pressures will continue to be measured with quartz gauges. Since hydrogen is a constituent of many spectroscopic additives, the Balmer spectrum will often be observed and will provide checks on the temperature and electron density via line intensity and line shape.^{19,20} As well as measuring

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atomic transition probabilities, we plan to deduce Stark-broadening parameters for those lines possessing measurable shapes and to investigate the sources of continuous spectra.

5. Acknowledgements

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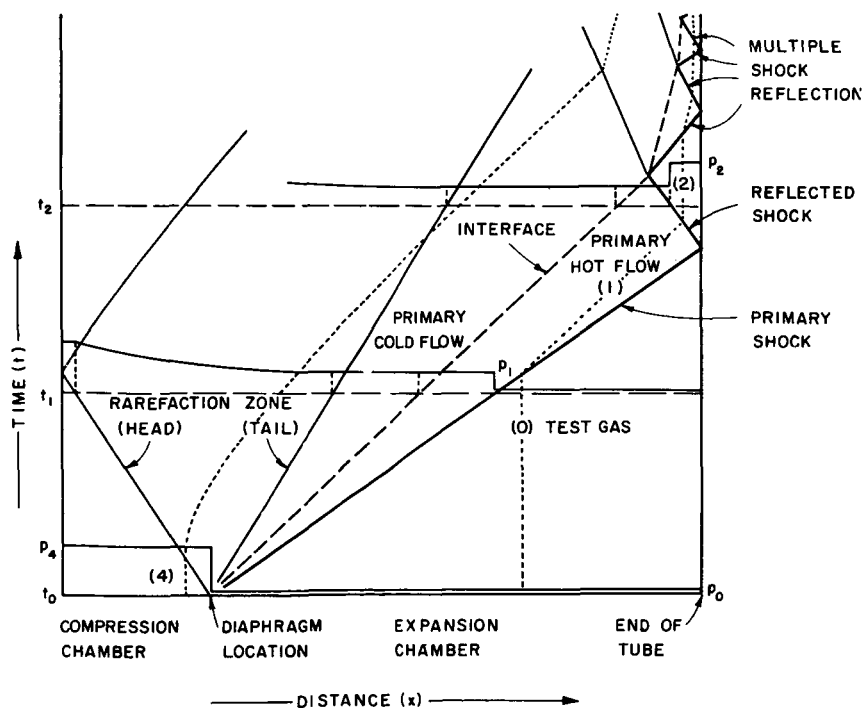


Figure 1. Space-time diagram of wave motion in conventional shock tube. The test gas (e.g., neon plus spectroscopic additive) is compressed by primary and reflected shock waves, and its emission spectrum is observed at far right.

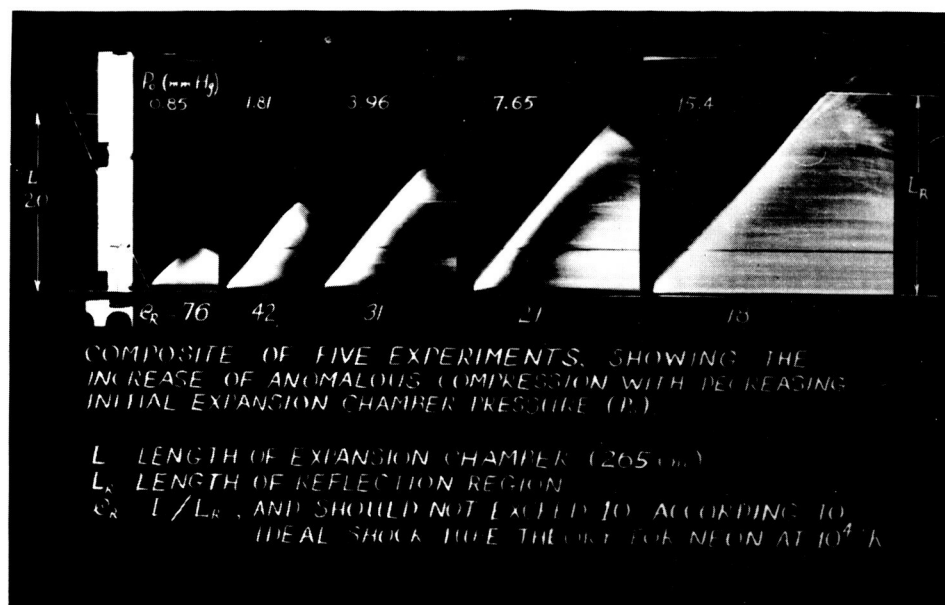


Figure 2. Shortening of flow column as function of initial pressure P_0 . Growth of boundary layers causes decreasing observing time with decreasing P_0 .

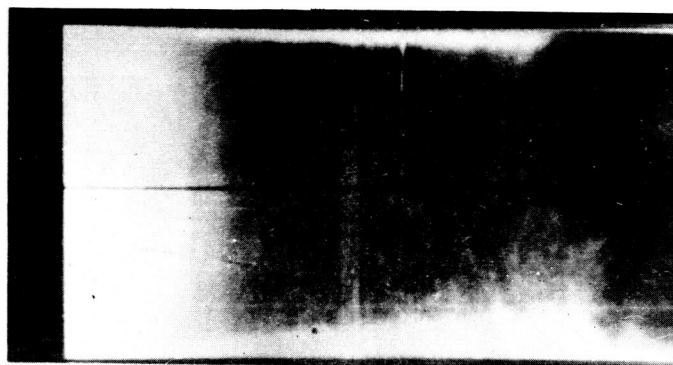


Figure 3. Transient C_2 emission in the primary flow, induced by addition of C_2H_2 to neon. Luminosity shows plane shock followed by turbulent boundary layers on tube walls.

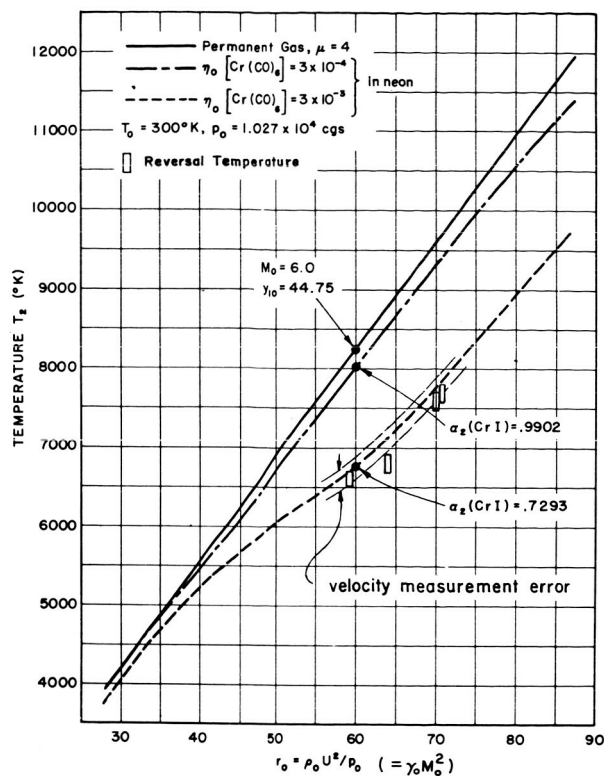


Figure 4. Theory and experiment for temperature behind reflected shock (T_2) as function of primary shock strength. Line-reversal temperatures confirm predictions for 3/10 % chromium carbonyl in neon.

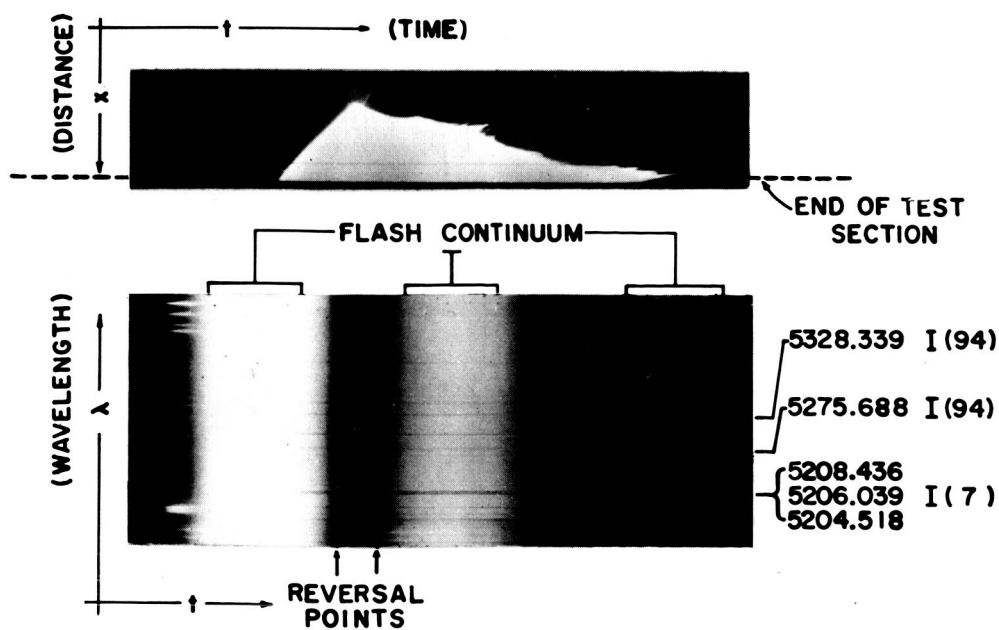


Figure 5. Composite of space-time photograph and time-resolved spectrogram for line-reversal experiment on chromium lines. Predicted: $T_2 = 6720 \pm 105^\circ\text{K}$; Measured: $T_2 = 6599 \pm 112^\circ\text{K}$ and $6473 \pm 145^\circ\text{K}$ on first and second reversals.

DISCUSSION

Question by R. S. BERRY (U.S.A.) :

Will you be able to carry your intensity measurements to the limit of the quartz region ?

Answer by G. CHARATIS (U.S.A.) :

Yes, we will be able to carry these temperatures hopefully down to 2000 Å by means of quartz windows.